Part II: A Novel Controller

By the use of an imperfectly mixed model developed for LDPE vessel reactors, we show that a linear controller cannot perform satisfactorily in the entire range of operating conditions of industrial interest. Based on the concept of reaction rate control, a novel controller is developed which provides satisfactory closed-loop dynamics independent of steady-state conditions. A nonlinear controller is shown to be more effective in avoiding reactor light-off.

SCOPE

The control of the low-density polyethylene (LDPE) vessel reactors is extremely important for an economic operation of the process. In industrial practice, the conttroller loop is designed to keep the polymerization temperature at the desired value and acts on the initiator feed rate. Technical problems arise not only from the particular dangers involved by the reacting fluid and in the very high operating pressure and temperatures, but also from the necessity of changing the polymerization conditions to change the quality of polymer product. Sometimes, the unsatisfactory performance of the external controller is an obstacle in obtaining the desired polymerization conditions, or it forces plant operators to undesired and time-wasting changes in the operating conditions. Earlier efforts in the analysis of the steady-state behavior and the dynamic characteristics of this high-pressure polyethylene process led to the development of an imperfectly mixed model (Marini and Georgakis, 1984), which was shown to be in very good agreement with most of the available experimental data. In particular, it was demonstrated that, in the region of industrial operating conditions, the reactor exhibits multiple steady states. The one of interest is either open-loop unstable or very close to that.

In this paper, we will use this model to design a satisfactory controller for LDPE vessel reactors. In particular, it will be shown that the open-loop dynamics of the reactor that become unstable are related to the deviation of the reaction rate from its steady-state value. By use of the concept of "reaction rate control" a novel controller is designed which performs satisfactorily throughout the range of operating conditions of industrial interest.

CONCLUSIONS AND SIGNIFICANCE

In the temperature range of industrial interest, the imperfectly-mixed low-density polyethylene vessel reactor effectively behaves as a first-order system. However, its open-loop dynamics are such that both the steady-state gain between initiator flow and reactor temperature and the process time constant drastically change magnitude and sign. For this reason, a linear PI controller, tuned at one operating temperature, does not perform satisfactorily at another one. A continuous tuning of the controller is thus required when the polymerization temperature is changed.

To design a more effective controller, we have shown that the unstable dynamics of the reactor are related to the deviation of the overall reaction rate from its steady-state value. This implies that the control loop design should be based on the concept of "Reaction Rate Control." A linear controller, with a proportional gain and an integral time explicitly dependent, as implied by the concept of reaction rate control, on the steady-state temperature, succeeds in providing satisfactory performances throughout the entire range of industrial operation. In fact, the closed-loop eigenvalues in the temperature range of interest becomes practically independent on the steady-state conditions. A nonlinear reaction rate controller is shown to be more effective than the linear one in avoiding reactor light-off when process disturbances cause significant decreases in the reactor temperature.

INTRODUCTION

Low Density Polyethylene is industrially produced in vessel reactors at very high pressures (1,000 ÷ 2,500 atm). The polymerization temperature ranges somewhere between 150 and 300°C, depending on quality considerations. To meet market demand, plant operators change the operating conditions of the reactor, such as polymerization temperature and pressure, and the type of initiator used. For this reason, modern vessel reactors are designed for a broad range of operating conditions. Because changes in the polymerization temperature have a direct influence on polymer quality, good temperature control is necessary for the reactor operation. Furthermore, there exists experimental evidence (van der Molen, 1981) that under certain conditions the reactor exhibits a tendency to extinguish the polymerization reaction. In industrial practice, the control of the polymerization temperature is generally performed by manipulation of the initiator feed rate. This has direct influence on the polymerization reaction, because the initiator feed decomposes and generates free radicals, which start to polymerize the monomer. Any change in the initiator feed has direct influence on the polymerization rate and on the heat production.

In Part I, our attention focused on the steady-state and stability characteristics of the LDPE reactor and led to the development of an "imperfectly mixed" model, which was shown to be in very good agreement with most of the available experimental data. In this paper, we will use this model for a discussion of the problems involved in the design of the reactor control system. The fundamental ideas and the basic equations of the model have been discussed in Part I. As a reference case, we have chosen a single-stage reactor which operates at polymerization temperatures in the range

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TABLE 1. MODEL PARAMETERS

Reactor Conditions

τ = residence time = 26.9 (s)
T^* = reference temperature = 180 (°C)
I_R^* = reference initiator feed = 36.74 (mmol/h)
T_o = input temperature = 70°C
P = operating pressure = 1,570 (atm)
M_o^* = input monomer concentration = 18.71 (mol/LT)

Kinetic Parameters

initiator #10:	$Da_I = 97.468$;	$\gamma_I = 35.011$;	$f = 0.6$
initiator #12:	$Da_I = 20.719$;	$\gamma_I = 35.90$;	$f = 0.98$
propagation:	$Da_P = 278.05$;	$\gamma_P = 8.084$	
termination:	$Da_T = 1.18 \cdot 10^9$;	$\gamma_T = 1.115$	

$$\beta = \frac{(-\Delta H)M_o^*}{\rho \cdot C_p \cdot T^*} = 2.942$$

Fluid-Mechanic Parameters

$$\alpha = \frac{Q^1}{Q^1 + Q^2} = 0.5 \qquad \xi_1 = \frac{V_1}{V} = 0.015$$

$$\epsilon = \frac{Q_r}{Q^1 + Q^2} = 3.75 \qquad \xi_2 = \frac{V_2}{V} = 0.015$$

$$\xi_3 = 1 - \xi_1 - \xi_2 = 0.97$$

of $160 \leq T \leq 250°C$. All the operating conditions, together with the model parameters and the kinetic constants, are given in Table 1. The nomenclature here is the same as in Part I.

PERFORMANCE OF CLASSICAL PI CONTROLLERS

In Part I, we demonstrated that the assumption of perfect mixing leads to incorrect predictions at high-temperature side of the operating region. However, the corresponding model is simpler and can be easily manipulated to provide information about the dynamic behavior of the reactor. For this reason, we first start by considering the reactor as perfectly mixed. The modifications introduced by imperfect mixing will be considered subsequently. The perfectly-mixed model is described, in dimensionless form, by Eqs. 5–8 of Part I. The complexity of this dynamic model can be reduced by employing the pseudosteady-state hypothesis with respect to the balances for the initiator and the radical species (Eqs. 5 and 6 of Part I). This hypothesis is justified by the fact that the eigenvalues related to these equations are always very small ($\cong -100, -1{,}000$) compared to the leading eigenvalue ($\cong 1$). Consequently, the intiator and radical concentrations, I and λ, can be calculated as function of temperature T and initiator feed I_o. The dynamic model is then represented by the following equations:

$$\frac{dM}{dt} = 1 - M - R \tag{1}$$

$$\frac{dT}{dt} = T_o - T + \beta R \tag{2}$$

where R indicates the overall polymerization rates, given by:

$$R = DaM \exp[\gamma(1 - 1/T)] \sqrt{I_o} \tag{3}$$

It is noted that R depends on the monomer concentration M, the polymerization temperature T, and the initiator feed I_o. The overall Damkholer number Da, and the dimensionless activation energy γ have the following expressions:

$$Da = Da_P \sqrt{2f/Da_T} \tag{4}$$

$$\gamma = \gamma_P - \gamma_T/2 \tag{5}$$

In deriving expressions 4 and 5, two further assumptions were made: a) the initiator decomposition rate is very high, $Da_I \exp[\gamma_I(1 - 1/T)] \gg 1$; and b) the radicals output from the reactor, λ, is negligible. To study the dynamics of the resulting system, it is necessary to linearize Eqs. 1 and 2 around the steady state. We introduce the following deviation variables:

$$x_1 = M - M_s; \quad x_2 = T - T_s; \quad U = I_o - I_{os}; \quad d = T_o - T_{os}$$

so that we can write, in matrix form:

$$\frac{dx}{dt} = Ax + BU + Cd \tag{6}$$

where

$$A = \begin{bmatrix} -1 - R_1 & R_2 \\ \beta R_1 & -1 + \beta R_2 \end{bmatrix} \quad B = \begin{bmatrix} R_3 \\ \beta R_3 \end{bmatrix} \quad C = \begin{bmatrix} 0 \\ 1 \end{bmatrix} \tag{7}$$

In expressions 7, we have indicated with R_1, R_2 and R_3 the steady-state sensitivities of the reaction rate R with respect of M, T and I_o respectively. Their values are known and depend only on the steady-state values of M, T and I_o. The linearized model (Eq. 6), transformed to the Laplace domain, produces the following transfer function relations:

$$\hat{x}_2(s) = G(s) \cdot \hat{U}(s) + G_d(s) \cdot \hat{d}(s) \tag{8}$$

with

$$G(s) = \frac{\beta R_3}{s + 1 + R_1 - \beta R_2} = \frac{A}{\tau_s s + 1} \tag{9}$$

This reactor model can now be seen as a first-order system. The open-loop pole, $-1 - R_1 + \beta R_2$ is positive and the steady-state gain $A = \beta R_3/(1 + R_1 - \beta R_2)$ is negative and not very sensitive on the reactor temperature. Even though the process is calculated to be open-loop unstable, it is possible to design a linear PI controller with satisfactory performances in a large range of polymerization conditions. For example, when initiator #10 is used in the usual temperature range ($T = 160 \div 200°C$), the open-loop characteristics are: $\tau_s \cong -1.2$ and $A \cong -0.18$. In this situation, a reasonable choice for the gain and integral time of a linear PI controller is: $K_c = 16$, $\tau_i = 4$. This choice would generate two equal closed-loop eigenvalues $s_1 \cong s_2 \cong -0.8$, which ensure a sufficiently fast response throughout the entire range of polymerization conditions.

This situation changes drastically when the effect of imperfect mixing is considered. In this case, the reactor can be modelled by three CSTR's in series (see Figure 2b of Part I), which a certain recirulating flow rate between them. The first two volumes, V_1 and V_2, account for a very small part of the entire volume (3%) and represent the conditions at the initiator feed point. Also in this case, the open-loop dynamics are characterized by two leading eigenvalues, while all the others are smaller by at least two orders of magnitude. The two slow modes are identified with the overall heat and monomer balance on the reactor, which can be written in the form:

$$\frac{dM}{dt} = 1 - M - R \tag{10}$$

$$\frac{dT}{dt} = T_o - T + \beta R \tag{11}$$

where M and T are respectively the output monomer concentration and temperature, equal to M_3 and T_3 of Part I. These equations are formally identical to Eqs. 1 and 2, but now the overall reaction rate R is the sum of the contributions in the three volumes V_1, V_2, and V_3.

$$R = \sum_{i=1}^{3} Da_P \xi_i M_i \lambda_i \exp[\gamma_P(1 - 1/T_i)] \tag{12}$$

Since all the terms in this expression depend on M, T and I_o, we can write, in compact form:

$$R = R(T, M, I_o) \tag{13}$$

We can now repeat the same analysis previously done for the simple CSTR model, arriving at the same conclusion that the system can be considered first order. The transfer function is still given by Eq. 9. The fundamental difference between the two models is that, when imperfect mixing is considered, both the open-loop time constant, τ_s, and gain, A, drastically change magnitude in the temperature region of interest. Figure 1 shows the dependence of the steady-state gain A on polymerization temperature for two types of initiators (#10 and #12) as predicted by the imperfectly-mixed model. The temperature range can be clearly divided

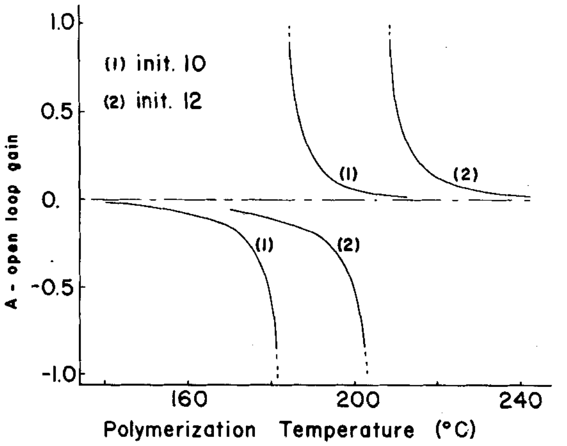

Figure 1. Open-loop gain of the imperfectly-mixed reactor as function of the polymerization temperatrure.

into two regions, one with negative and one with positive gains. Temperature A also exists where the gain is infinite. In curve 1 of Figure 2 we present the temperature dependence of the open-loop eigenvalue ($-1/\tau_s$). We clearly see that at low temperatures the reactor is open-loop unstable and that it quickly becomes open-loop stable at high temperatures. With these open-loop characteristics it is not possible to tune a classical linear controller at one temperature and expect it to perform satisfactorily over the entire temperature range of interest. Figure 2 also shows the temperature dependence of the leading closed-loop eigenvalue, generated by the use of classical linear PI controllers. The continuous line represents real eigenvalues while the broken line indicates the real part of a pair of complex conjugate eigenvalues. The reactor is operated at the reference conditions of Table 1 with initiator #12.

Two such classical controllers are examined. They both have constant proportional band and integral time. The first controller (curve 2) is tuned at a low temperature (T = 190°C), and it performs satisfactorily in this region, since the closed-loop eigenvalue is of the order of −1. On the contrary, when the polymerization temperature is increased, two real eigenvalues are generated, one of which becomes very small (≤0.1) indicating unsatisfactory slow dynamics. The second controller (curve 3) is tuned at a high temperature (T = 225°C), where its performances are very good. When the temperature is decreased, two complex eigenvalues are generated. The absolute value of their real part decreases with temperature while the imaginary part increases. This gives origin to high oscillations. At lower temperatures the reactor becomes unstable prohibiting reactor operation at the temperature range of 160–190°C. The situation described by Figure 2 reflects very well the different control requirements of the reactor at different operating conditions. In fact, at low temperature the open-loop unstable reactor needs a high controller gain to stabilize the system, while a relatively low integral action is needed to compensate for load disturbances. On the contrary, at high temperatures the system is already stable and needs a low gain and a relatively high integral action. This implies that the classical linear PI controller must be separately tuned at each polymerization temperature. Furthermore a change in the initiator type requires a change in the controller parameters, even though the reactor might operate at the same polymerization temperature (see Figures 1 and 6 of Part I).

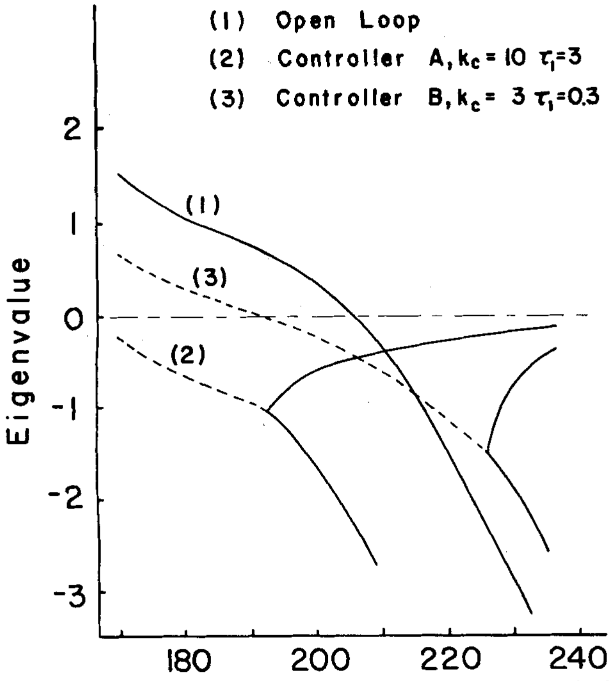

Figure 2. Leading eigenvalue of the system as function of the polymerization temperature, for initiator #12; (——) indicates a real eigenvalue (- - -) indicates the real part of two complex conjugates eigenvalues.

REACTION RATE DYNAMICS

The linearized dynamics as expressed by Eqs. 1 and 2 lead to the calculation of the following two eigenvalues: $\lambda_1 = -1$, $\lambda_2 = \beta R_2 - R_1 - 1$. The corresponding eigenrows are

$$V_1^T = (\beta,1)$$

$$V_2^T = (R_1,R_2) \tag{14}$$

The first eigenvalue is related to the adiabatic nature of the reactor. This is also apparent from the corresponding eigenrow. Obviously, the corresponding dynamic mode cannot be controlled by manipulating the initiator feed rate. Even further, this mode need not be controlled because the corresponding eigenvalue, $s_1 = -1$, is constant and quite negative. Focusing on the second eigenvalue which is unstable, one notices from Eq. 14b that the dynamic mode is the linearized form of the deviation of the reaction rate, $R(M,T)$, from its steady-state value $R_s = R(M_s,T_s)$. That is:

$$z_2 = v_2^T x = R_1 \delta M + R_2 \delta T \doteq R(M,T) - R(M_s,T_s)$$

We will now argue that, with some approximation, the same conclusion can be reached with the imperfectly-mixed model. Specifically, the dynamic mode corresponding to the eigenvalue that become open-loop unstable is related to the deviation of the overall polymerization rate from its steady-state value. To assess the magnitude of the approximation we calculate how closely the following (1 × 12) row:

$$h^T = (0,0,0,0,0,0,0,0,0,0,R_1,R_2)$$

represents the eigenrow v^T that corresponds to the unstable eigenvalue of the system of Eqs. 14–25 of Part I. Here R is given by Eq. 12, and R_1 and R_2 denote sensitivities with respect to M_3 and T_3 of Part I, respectively. The angle defined by:

$$\cos\theta = \frac{h^T v}{\|h\| \cdot \|v\|}$$

was calculated for initiators #10 and #12 throughout the temperature region of industrial interest. Its value was found to be always very small ranging between 4 and 15 degrees. This proves that the dominant open-loop dynamic characteristics of the reactor are very closely approximated by the deviation of the overall reaction rate from its steady-state value.

REACTION RATE CONTROLLER

We showed that the unstable dynamics of the LDPE vessel reactor are associated with the deviation from steady state of the reaction rate R. It was also discussed that the reaction rate is directly influenced by M, T and I. Consequently the ideal nonlinear controller which adjusts I to keep the reaction rate at its steady-state value can be expressed by:

$$R(M(t),T(t),I_o(t)) = R(M_s,T_s,I_{os})$$

from which $I_o(t)$ should be solved either explicitly or implicitly in the form:

$$I(t) = G(M(t),T(t);I_{os},M_s,T_s)$$

With such a controller in action the closed-loop dynamics of the system are described by:

$$\frac{dx_1}{dt} = -x_1$$

$$\frac{dx_2}{dt} = -x_2$$

The above equations are obtained without the need of any linearization and thus are valid globally, for any magnitude of the perturbation variables x_1 and x_2.

Consequently, the reactor is always closed-loop stable, and the closed-loop eigenvalues are equal and corresponds to a dimensional closed-loop time constant of one residence time.

A linearized version of this controller is obtained by writing

$$\delta R = R_1x_1 + R_2x_2 + R_3U = 0$$

from which we readily derive that

$$V = -\left(\frac{R_1}{R_3}x_1 + \frac{R_2}{R_3}x_2\right) \tag{15}$$

which represents the ideal linearized "Reaction Rate" controller. It requires the measurement of the monomer concentration deviation (x_1) and the polymerization temperature deviation (x_2) from their steady-state values, and the calculation of the reaction rate sensitivities, R_1, R_2, and R_3, as functions of T_s,M_s and I_o.

Several practial problems arise at the implementation of such a controller. First, the on-line accurate measurement of the monomer concentration is not easy. We can overcome this by assuming that the total enthalpy, $\beta M + T$, of the reactor is always at steady state. This leads to:

$$x_1 = -x_2/\beta \tag{16}$$

Secondly, we need an appropriate expression for the overall reaction rate $R = R(T,M,I_o)$ to calculate R_1, R_2 and R_3. With respect to the imperfectly-mixed model which accurately represents the reactor behavior, we can derive an approximate expression by the use of the following reasonable assumptions:

1) The monomer concentration in the three volumes, V_1, V_2 and V_3, is practically the same: $M_1 \cong M_2 \cong M_3 = M$.

2) The contribution of the two small volumes, V_1 and V_2, to the overall reaction rate R in Eq. 12 is negligible.

3) In each volume, the termination rate for the radicals is equal to the production rate (quasisteady-state assumption).

The following approximate expression is then derived:

$$R = M \cdot F(T) \cdot \sqrt{I_o} \tag{17}$$

$$F(T) = Da\ \exp[\gamma(1 - 1/T)]\sqrt{r_i/(1 + r_i + \alpha r_i^2)} \tag{18}$$

In expression 18 Da and γ are still given by Eqs. 4 and 5; r_i is the initiator decomposition rate:

$$r_i = Da_I \exp(\gamma_I - \gamma_I/T)$$

and α takes into account the fluid dynamic characteristics of the reactor model:

$$\alpha = 0.4\frac{\tau_1 + \tau_2}{\tau} = 0.4\frac{\text{mixing time}}{\text{residence time}}$$

Here τ_1 and τ_2 are, respectively, the residence times in the first two volumes, V_1 and V_2, and the constant 0.4 takes into account the fact that in these volumes the reaction temperature is lower than in the bulk of the reactor. Then Eq. 15 becomes:

$$U = -K_c \cdot x_2$$

$$K_c = +2I_s\left[\frac{F'(T_s)}{F(T_s)} - \frac{1}{\beta M_s}\right] \tag{19}$$

In essence, the above equation represents a linear controller, but with the proportional gain K_c dependent on the steady-state conditions, in accordance with the concept of reaction rate control. With the influence of the proportional control defined by expressions 19 the closed-loop eigenvalue of the reactor is approximately equal to -1 throughout a large temperature range of practical interest. This proportional "Reaction Rate" controller takes into account and approximately compensates for the changes in the open-loop dynamics of the reactor.

For a successful application, the controller should also be able to handle load disturbances. This requires the introduction of integral action in the controller of Eq. 19 in the following form:

$$U = K_c(T_s - T) + K_I\int(T_s - T)dt \tag{20}$$

where the gain K_c is defined by Eq. 19 and the constant of the integral action is given by:

$$1/K_I = 4\beta R_3 = 2\beta\frac{1 - M_s}{I_s} \tag{21}$$

The value of K_I is chosen to obtain two real closed-loop eigenvalues equal to -0.5. This avoids oscillations and maintains a sufficiently fast response.

For a practical application, expression 20 can be simplified by eliminating some terms, as M_s, which change very slowly with polymerization conditions. The final form of the linearized version of the reaction rate controller is

$$U = 2I_s\left[\left(\frac{F'(T_s)}{F(T_s)} + K_c'\right)(T_s - T) + \frac{K_c'}{\tau_I'}\int(T_s - T)dt\right] \tag{22}$$

with

$$\frac{F'(T_s)}{F(T_s)} = \frac{\gamma}{T_s^2} + \frac{\gamma_I}{T_s^2}\frac{1 - \alpha r_i^2}{2(1 + r_i + \alpha r_i^2)} \tag{23}$$

and the corresponding scheme is represented in Figure 3. In expression 22 we have introduced two constants K_c' and τ_i', which will play a similar role as their usual counterparts play in the classical PI controllers. They appear in the terms of the reaction rate controller where a dependence on M_s existed. These parameters can be determined on-line or can be calculated by the use of the reactor model so that the close-loop eigenvalues have a desired value. The terms $F'(T_s)/F(T_s)$ accounts for the necessity of varying the overall gain with the polymerization conditions. The $2I_s$ term accounts for the sensitivity of both the proportional and integral action with the initiator feed.

Figure 4 shows the closed-loop eigenvalues of the system, with the reaction rate controller defined by Eq. 22, as function of the polymerization temperature. Three choices of the two constants K_c' and τ_i' are examined. It can be seen that a satisfactory closed-loop behavior is obtained in all cases. In the temperature region of interest, the closed-loop system is characterized by two real eigenvalues that are approximately equal to each other (part B-C of curve 2) and their value varies very little with the polymerization temperature. At low temperatures ($T < 180°$C) two complex ei-

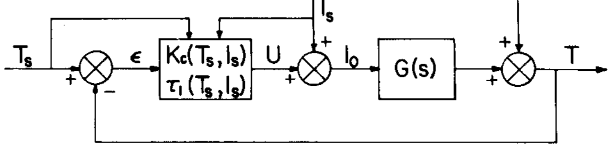

Figure 3. Schematic representation of the linear reaction rate controller.

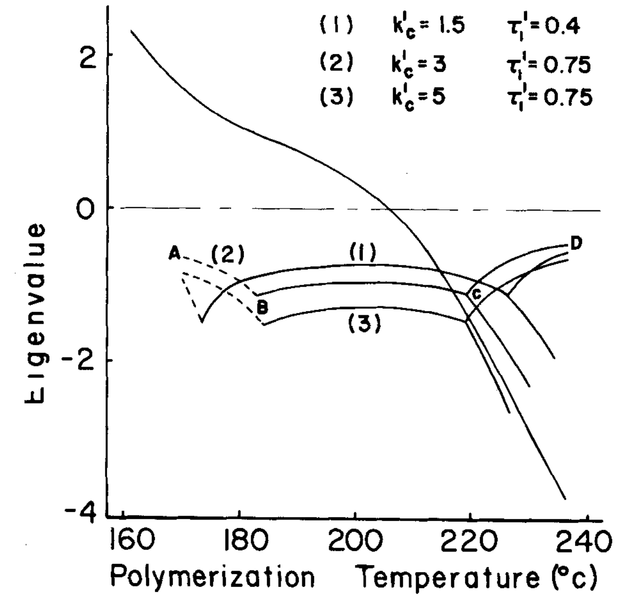

Figure 4. Leading eigenvalue of the system as function of the polymerization temperature, for three different reaction rate controllers. Initiator #12.

genvalues are generated (part A-B of curve 2), where we only plot the real part. The imaginary part increases with decreasing temperature. Now the reactor can no longer be represented by Eqs. 10 and 11 only. The transient term of the initiator mass balance must be taken into account because the initiator decomposition is no longer very fast. On the other hand, at high temperatures ($T >$ 225°C) two different real eigenvalues are generated (part C-D of curve 2), and one of them decreases with increasing temperature. At these temperatures, expression 17 fails to give a good approximation of the overall reaction rate. Both temperature regions where the controller does not behave as ideally as desired are not of industrial interest. In fact, at low polymerization temperatures the point of light-off temperature is approached, where as shown in the Appendix the reactor is always closed-loop unstable. On the other hand, at high temperatures the initiator consumption is so high that one changes to another initiator type.

To successfully apply the reaction rate controller to the industrial process, the value of the following two parameters are required: γ, the overall activation energy of the reaction, and r_i, the initiator reaction rate. Because their values are known approximately, it is important to inquire whether the controller performance depends on the accuracy with which these parameters are known. For this reason we performed calculations of the closed-loop eigenvalues using in the controller algorithm values for γ or Da_I that were different from those used in the mathematical model that represents the real reactor. According to the general knowledge of this polymerization reaction, the value of γ was varied in the range ±20%, while the initiator Damkholer number, Da_I, was varied ±100%. In all cases, the satisfactory performance of the closed-loop eigenvalues of Figure 4 was retained. This indicates that the controller performance is not dependent on a perfect knowledge of the reaction parameters. Calculations with other types of initiators showed that the reaction rate controller designed performs very satisfactorily over the entire range of polymerization temperature ($150 \leq T \leq 260$°C), and without any need to update the parameters K_c' and τ_i' for the different initiators.

STEP RESPONSE OF CLOSED-LOOP SYSTEM

The analysis carried out in the previous sections was based on the linearized version of the equations that describe the LDPE vessel reactor. However, the nonlinearities of the real system may influence the dynamics of the reactor. For this reason it is important to verify the performances of the controllers designed by simulation of the reactor through model B. The various controllers tested are: "controller A," the classical linear controller tuned at low temperature (Figure 2, curve 2); "controller B," the classical linear one tuned at high temperature (Figure 2, curve 3); and "controller C," the reaction rate controller defined by Eq. 22 (Figure 4, curve 2).

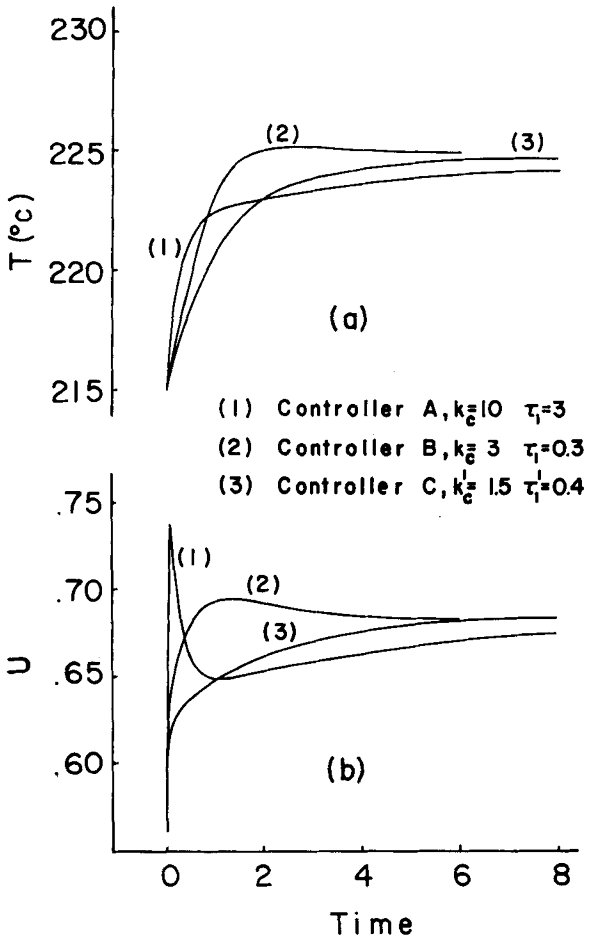

Figure 5. System response to a step change of the polymerization temperature set point from 215 to 225°C. Initiator #12.

Figure 5 shows the reactor temperature response to a step change in the reactor temperature set point from 215 to 225°C. As expected, both controllers B and C produce a satisfactory response: the reactor reaches the new steady state after approximately four residence times (nearly 2 min). On the contrary, controller A, which was tuned at low temperature, gives origin to a slow approach to the steady state. Furthermore, controller A produces an unsatisfactory large change in the initiator feed (Figure 5, curve 2), which is first increased over the final value, then decreased, and finally increased very slowly to its new steady-state value. The initial very fast increase in the initiator feed may have very dangerous consequences on the operation of the polyethylene reactor.

Figure 6 shows the response of the system to a step change of the feed temperature from 70 to 60°C when the reactor operates at 195°C. It is noted that in this case both controllers A and C produce a satisfactory response. On the contrary, controller B, which was tuned at a high temperature, is practically unable to control the reactor and gives origin to strong oscillations which finally lead to instability.

The above examples confirm the conclusions obtained from the linear analysis of the system. The reaction rates controller produces good dynamic response in a large temperature range, while the classical linear controller needs tuning at each polymerization temperature. The performance of the controller becomes quite crucial when the reactor is operated at a very low temperature. In

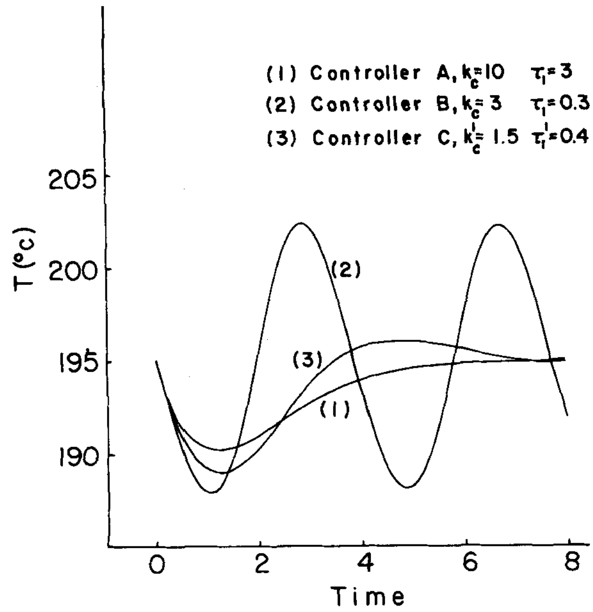

Figure 6. System response to a step change of the feed temperature from 70 to 60°C. Initiator #12; temperature set point = 195°C.

the Appendix we show that for each initiator there exists a minimum temperature, the light-off temperature, under which the reactor becomes unstable, independent of the controller. However, when the reactor is operated at a temperature not too far from this minimum value, local stability is not sufficient to ensure the satisfactory operation of reactor.

Figure 7 shows transients of the reactor operating at a temperature of 180°C with initiator #12. This temperature is sufficiently far from the minimum "light-off" temperature, which is approximately equal to 158°C. From the linear analysis we expect controller A to perform satisfactorily, since the leading closed-loop eigenvalue is equal to −0.7 (Figure 2, curve 2). Indeed, when the input feed temperature is changed from 70 to 60°C, controller A is not able to keep the polymerization conditions, and the reactor becomes extinct (Figure 7, curve 1). On the contrary, controller C, based on the concept of reaction rate control, succeeds in maintaining the polymerization conditions. This is due to the fact that the controller parameters, gain and integral time, are self-tuned with respect to the polymerization conditions.

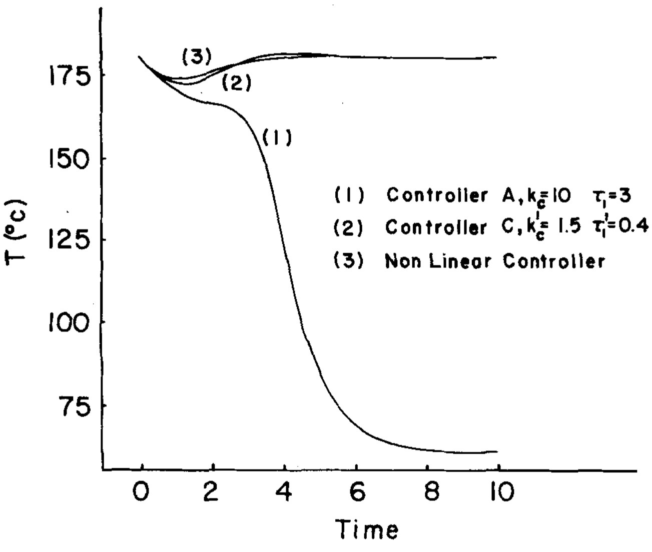

Figure 7. System response to a step change of the feed temperature from 70 to 60°C. Initiator #12; temperature set point = 180°C. Maximum initiator feed equal to two times the reference one.

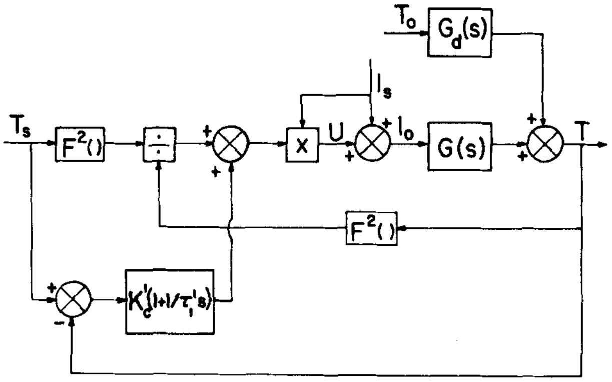

Figure 8. Schematic representation of the nonlinear reaction rate controller.

In these critical situations better performances can be obtained by using a nonlinear reaction rate controller schematically shown in Figure 8. It is divided into two parts. The bottom one represents the linear part of the controller that includes integral action. The top part takes into account the nonlinearities of the reactor by using the same function $F(T)$ defined by Eq. 18. If linearized this nonlinear controller reduces to the one defined by Eq. 22. The major advantage of this nonlinear controller is that it automatically modifies the overall proportional gain with the size of the temperature deviation and provides large gains for large deviations and small gains for small deviations. In Figure 7 (curve 3), the performance of this nonlinear controller is shown to be slightly better than that of its linear version. In other noncritical situations this controller has the same performances with its linearized version.

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APPENDIX: CALCULATION OF MINIMUM LIGHT-OFF TEMPERATURE

When the polymerization temperature is sufficiently low, we can consider the entire reactor as perfectly mixed. In this region of operation, both the steady-state and the stability characteristics of the reactor are very well represented by the simple CSTR model (Marini and Georgakis, 1982, Figures 3 and 4). Since the related eigenvalue is very low (≅ −1,000), we will make the approximation that the radicals are at steady state, which leads to

$$\lambda = \left(\frac{2fIDa_I(\gamma_I - \gamma_I/T)}{Da_T \exp(\gamma_T - \gamma_T/T)}\right)^{1/2}$$

In this situation, the reactor behavior is described by the following equations:

$$\frac{dI}{dt} = I_o - I - R_1 \tag{A1}$$

$$\frac{dM}{dt} = 1 - M - R_2\sqrt{R_1} \tag{A2}$$

$$\frac{dT}{dt} = T_o - T + \beta R_2\sqrt{R_1} \tag{A3}$$

where:

$$R_1 = Da_I \exp\left[\gamma_I\left(1 - \frac{1}{T}\right)\right]$$

$$R_2 = Da_p\left(\frac{2f}{Da_T}\right)^{1/2} \exp\left[\gamma\left(1 - \frac{1}{T}\right)\right] M$$

We now introduce the following deviation variables:

$$x_1 = I - I_s; \quad x_2 = M - M_s; \quad x_3 = T - T_3; \quad U = I_o - I_{os}$$

and linearize the system around the steady state. If the control variabile U is related to the polymerization temperature x_3 in the form of Eq. 19

$$U = -K_c \cdot x_3 \qquad \text{(A4)}$$

the characteristic equation of system (Eq. A4), in the Laplace domain, is in the form:

$$(s + 1)(s^2 + As + B) = 0 \qquad \text{(A5)}$$

It is simple to verify that the term A does not contain the controller gain K_c. This term is equal to:

$$A = D_{aI} \exp\left[\gamma_I\left(1 - \frac{1}{T_s}\right)\right] + (T_s - T_o) \times \left[\frac{1}{M_s} - \frac{\gamma}{T_s^2} - \frac{1}{2}\frac{\gamma_I}{T_s^2}\right] + 2 \qquad \text{(A6)}$$

When $A < 0$, the system is always unstable because Eq. A5 has always a positive root. The value of T_s for which $A = 0$ defines the minimum light-off temperature that is the critical value under which the reactor is always closed-loop unstable, for any choice of the controller parameters. In the reference conditions (Table 1), this temperature is equal to 137°C for initiator #10 and to 158°C for initiator #12. From an analysis of Eq. A6 it is noted that, for a given initiator, the minimum light-off temperature is decreased only by increasing the input temperature T_o.

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Finishing Stages of PET Synthesis: a Comprehensive Model

A comprehensive mathematical model for the finishing stages of polyethylene terephthalate (PET) synthesis is developed. The model takes into account the variables which have significant influences on polymerization but have not been hitherto taken into account, e.g., changing interfacial concentration and the presence of side reactions. The model is applicable to industrial polymerizations, and the predictions agree well with the literature and with certain pilot-plant and commercial data.

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SCOPE

During the manufacture of polyethylene terephthalate, a complex series of reactions accompanies the main polycondensation reaction. Apart from playing an important role in controlling the product quality (such as fiber color, dyeability and stability), these side reactions also modify the progress of polymerization. The modelling problem, however, is formidable due to the coupling of the diffusional and kinetic phenomena occurring in the final stages.

There have been some efforts to model this process in the past. However, in all these cases, simple approximations, such as the neglect of the presence of side reactions and assumption of constant interfacial concentration, have been made. Such simplified analyses, although valuable from the point of view of providing first insights, are not directly pertinent as far as the industrial reactor operations are concerned. A need to develop a more comprehensive model which avoids these deficiencies was felt. Since PET is a large-tonnage polymer, any insight into the complex interactions in the finishing stages leading to improved methods of design, productivity, and product quality control is extremely useful.

CONCLUSIONS AND SIGNIFICANCE

A comprehensive model incorporating the major side reactions that occur during the PET formation and also accounting for the continuous change of the interfacial concentration of the volatile species was developed. An approximate numerical technique was devised to calculate the interfacial concentration.

The influence of important process and operational variables, such as pressure, temperature, mixing and film thickness (equivalent to the reciprocal of specific interfacial area), was examined; the trends predicted by the model were successfully compared with the available experimental and industrial data. Some novel and "unobvious" predictions emerged from the